

Wastewater irrigation beneath the water table: analytical model of crop contamination risks

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ABSTRACT

Wastewater irrigation alleviates freshwater scarcity. However, conventional (near)surface irrigation techniques directly expose crops to contaminants. Irrigating wastewater into shallow phreatic zones to raise the water table enhances groundwater evapotranspiration, while using the vadose zone as a bioreactor that attenuates contaminants through dilution, adsorption, and biodegradation. Nevertheless, contaminants may spread across the groundwater, soil, and vegetation. In this study, we focus on the crop contamination risks, and derive a simple analytical model to estimate crop solute uptake. Although crops are not directly exposed to the irrigated wastewater, contaminants (and nutrients) may spread to the root zone. Results show that crop contamination is primarily determined by the root zone water balance, and by solute dispersion and biogeochemical reaction parameters. The model contributes towards identifying hydrogeologically and climatically suitable locations for phreatic zone wastewater irrigation, determining acceptable levels of irrigation water quality, and evaluating crop contamination hazards against the fertigrative value of wastewater.

1. Introduction

Freshwater is a crucially important resource, and climate change is increasingly threatening freshwater availability (Schewe et al., 2014) and crop growth (Jiao et al., 2021) across the globe. As around 90% of global anthropogenic water consumption is used for irrigation, adapting agricultural practices towards the sustainable use of water resources alleviates freshwater scarcity, and mitigates the wider effects of climate change on food production and land degradation (Rosa, 2022; Konapala et al., 2020). Aside from freshwater scarcity, irrigation may also be constrained by water storage (Schmitt et al., 2022). Groundwater aquifers, in addition to their water storage capacities, are also important sources of water for agriculture in regions with shallow water tables. The large impacts of groundwater evapotranspiration (i.e., crop uptake of moisture originating from groundwater) on both crop yields and groundwater levels has led to a large ongoing research effort into optimizing groundwater evapotranspiration in agricultural systems (Hou et al., 2023). Another important source of water for agriculture is water stored in the vadose zone. Although frequently omitted from root zone water budget models, the recirculation of vadose zone moisture back into the root zone contributes substantially towards meeting crop water

requirements, and this is enhanced when groundwater resources are abundant and water tables are shallow (Kroes et al., 2018).

The development of new irrigation techniques that utilize water of marginal quality (e.g. treated wastewater), to conserve freshwater, is being stimulated through national and international policies (Narain-Ford et al., 2020). However, using wastewater for irrigation may lead to risks associated with the contamination of soil, groundwater, surface water, and crops (Maffettone and Gawlik, 2022). Crops are particularly at risk of contamination if the crops are directly exposed to the wastewater, for example with conventional (near)surface irrigation methods (Beggs et al., 2011). Wastewater suitable for irrigation typically contains low concentrations of organic contaminants (e.g. pharmaceuticals, hormones, household chemicals), which are collectively known as contaminants of emerging concern (CECs). Such organic contaminants may be highly amenable to attenuation by adsorption to the soil matrix, and biodegradation by soil organisms (Narain-Ford et al., 2022). Therefore, developing wastewater irrigation techniques that do not directly expose crops to contaminants is crucial for food safety and public acceptance (Verhoest et al., 2022).

Phreatic zone wastewater irrigation is being considered as a new method of managed aquifer recharge that replenishes groundwater

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aquifers, and functions as a technique for agricultural irrigation and additional wastewater treatment (Narain-Ford et al., 2022; Tang et al., 2023). With this water management strategy, (treated) wastewater is directly pumped into the phreatic zone through buried pipes, which recharges the groundwater, raises the water table, and increases capillary fluxes of groundwater and vadose zone moisture to the root zone (Fig. 1a). The vadose zone soil between the water table and the root zone, facilitates and provides time and space for contaminant attenuation processes. Two key pathways of crop contamination are mitigated: root uptake of contaminants, and contaminant attachment to the surfaces of edible portions of the crops (Partyka and Bond, 2022; Troldborg et al., 2017).

Phreatic zone wastewater irrigation also facilitates the reuse of water resources that might otherwise go to waste, while making more efficient utilization of existing agricultural infrastructure. Densely populated regions are especially susceptible to exacerbating freshwater scarcity, due to growing populations and increasing urbanization (He et al., 2021), yet urban regions also produce large amounts of treated wastewater that would be disposed of in surface water channels if not used for irrigation (Beard et al., 2019). Although discharging wastewater already fulfils a function in regional water management, for instance to prevent streams from falling dry during the summer (Pronk et al., 2021), exploiting treated wastewater for agriculture around urban areas represents a large untapped potential for water reuse and sustainability (Minhas et al., 2022). Within this framework, phreatic zone wastewater irrigation might limit crop contamination risks. Around 34% of agricultural land in the Netherlands is already underlain by drainage pipes, originally laid to mitigate water excess, that may be used for phreatic zone wastewater irrigation (De Wit et al., 2022). In areas with a wastewater treatment plant, wastewater could be considered as water supply source (Narain-Ford et al., 2021). These drains can simultaneously be used for controlled drainage to prevent waterlogging (de Wit

et al., 2024) and reduce nutrient leaching (Bonaiti and Borin, 2010), by adaptively reversing the direction of flow from irrigation to drainage if precipitation events raise water levels too much.

In order to support the responsible and safe implementation of phreatic zone irrigation with wastewater, we seek to develop a simple one-dimensional analytical model of crop contamination risk under phreatic zone wastewater irrigation, a new wastewater irrigation technique for which no analytical model currently exists. The main objective of this study is to build the analytical model, explain the physical processes that are considered in the derivation of the model, perform a sensitivity analysis of the model, and to validate it using data and insights from an existing field experiment and relatively complex two-dimensional numerical model. The analytical model may be used to coarsely estimate crop contamination risks and perform sensitivity analyses of crop contamination risks in response to variations in environmental, hydrological, and hydrogeological conditions, along with contaminant adsorption and biodegradation parameters. This enables the rapid identification of potential regions for implementing phreatic zone wastewater irrigation, possibly with large-scale geospatial data. In developing the model, we make conservative assumptions regarding contaminant transport behavior, and err on the side of overestimating crop contamination risks where necessary. This conservative approach allows us to make more conclusive comparisons against conventional (near)surface methods of irrigation, which directly expose crops to contaminants. The model will therefore be useful for designing and making regulatory guidelines for such phreatic zone irrigation systems, and determining permissible irrigation water quality in relation to crop contamination risks, from a risk-averse approach. With the aid of the model, we assess the merits, risks and implications of phreatic zone wastewater irrigation in the context of crop contamination, and discuss the possible fates of the contaminants in the wider environment, in relation to other methods of irrigation and wastewater disposal.

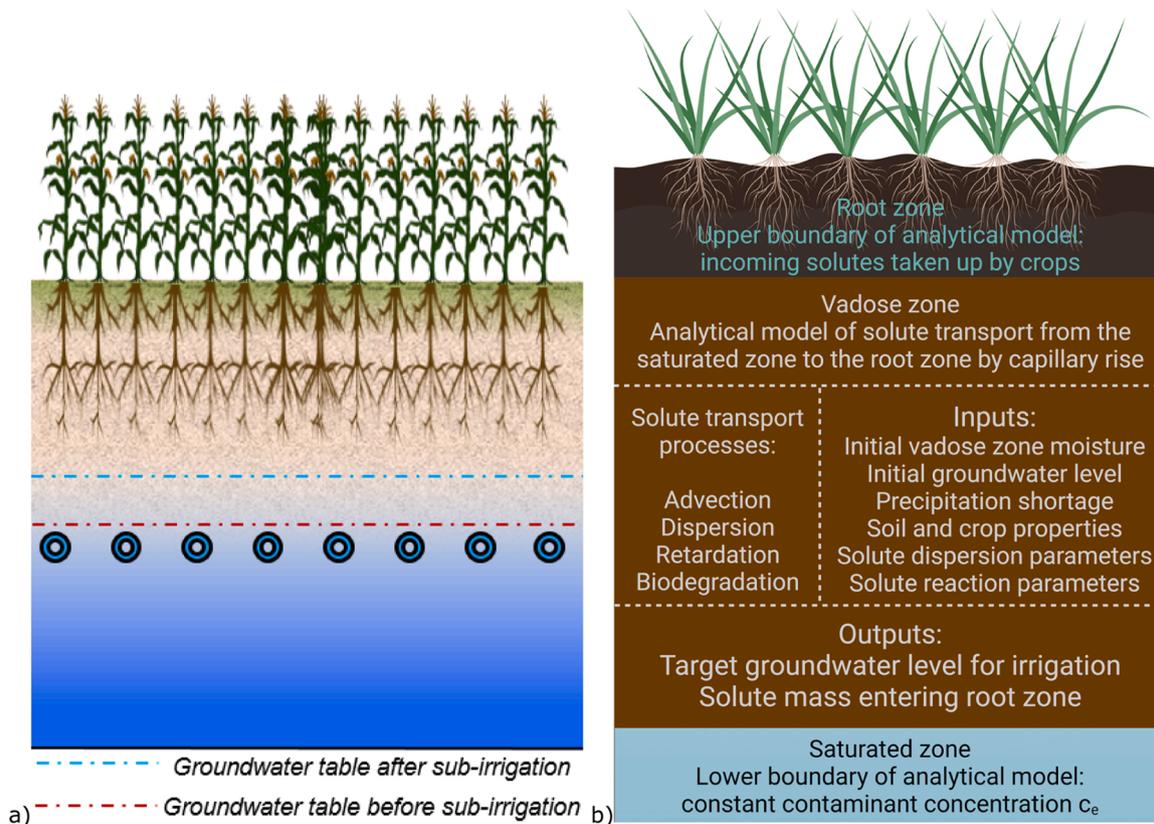


Fig. 1. (a) Conceptual illustration of the phreatic zone irrigation system. (b) Conceptual overview and illustration of the analytical model and a list of the processes it considers, with inputs and outputs.

2. Model development for crop contamination risk

2.1. Root zone water balance during the crop season

To minimize the risks of crop and root zone soil contamination, this new phreatic zone irrigation technique is recommended for regions with an annual precipitation excess, such as the Netherlands. Despite the annual precipitation excess, irrigation remains necessary during the crop season (e.g. April to September) because precipitation alone is insufficient to sustain optimal agricultural yields during the crop season. The annual precipitation excess, combined with the lack of wastewater irrigation outside the crop season, allows the vadose zone and underlying groundwater to be mostly flushed of CECs by infiltrating rainwater before the commencement of the following crop season (Narain-Ford et al., 2022). The risk of crop contamination with the new irrigation technique was found to be elevated if the pre-irrigation moisture content of the soil above the irrigation drains was low, or if drought occurs before or during a crop season (Tang et al., 2023). In these cases, it became necessary to irrigate larger volumes to compensate for deficits in rainfall or vadose zone soil moisture, thereby introducing more CECs into the soil. On the other hand, saturated zone hydraulic properties and their spatial heterogeneity were found to have minimal effect on crop contamination risks. Altogether, the solute mass balance in the vadose zone in each year is essentially independent of other years, and is primarily determined by the water balance in the vadose zone within individual crop seasons (Narain-Ford et al., 2022). Therefore, the risks of crop contamination due to CECs from phreatic zone wastewater irrigation may be effectively characterized with models that describe the water balance in the root zone, and solute transport in the vadose zone, during a single crop season.

To model crop contamination risks under phreatic zone wastewater irrigation, we first characterize the water balance in the vadose zone during the crop season. Subsequently, we model solute advection, dispersion, and biogeochemical reactions to describe its transport from the phreatic zone to the vadose zone, and root solute uptake. A comprehensive list of all model parameters and variables is given in Table 1. An overview and illustration of the processes, inputs, and outputs considered in the model is given in Fig. 1b. Three sources of water contribute to the total crop evapotranspiration volume over the crop season ET_c :

$$ET_c = P + S + G \quad (1)$$

where P is the total rainfall volume, S is the vadose zone moisture available for recirculation, and G is groundwater evapotranspiration. For simplicity, we assume that crop water requirements are satisfied from the three compartments in the order of preference $P \rightarrow S \rightarrow G$. The implications and limitations of this assumption are discussed later. Eq. 1 can be expressed in terms of the precipitation shortage

$$ET_c - P = S + G \quad (2)$$

which makes it evident that once the vadose zone available moisture S is depleted, the precipitation shortage must be filled by groundwater evapotranspiration for optimal crop yields to be maintained. Note that the volumes referred to here are for the crop season, approximately five months from April or May to August or September in our experimental site in the Netherlands (Narain-Ford et al., 2022), and not for the entire year.

2.2. Vadose zone soil water content before the crop season

We now calculate S , the vadose zone stored moisture content at the beginning of the crop season. This is the quantity of vadose zone water available to crops through recirculation after the water table is raised. The vadose zone soil moisture content is affected by atmospheric fluxes (rainfall and evapotranspiration) and the exchange of water with the

Table 1

Nomenclature. Symbols for units are L (length, mm), T (time, day), M (mass).

Alphabet	Units	Description
B	[T]	Empirical parameter in equation 37
c	[−]	Dimensionless solute concentration
c_b	[M/L]	Background solute concentration
c_e	[M/L]	Groundwater solute concentration
D	[L ² /T]	Hydrodynamic dispersion coefficient
e	[L/T]	Potential evapotranspiration rate
ET_c	[L]	Crop evapotranspiration volume
ET_{ref}	[L]	Reference evapotranspiration volume
f	[−]	Crop factor
F	[L/T]	Irrigation flux required to maintain target groundwater level
G	[L]	Crop water uptake originating from groundwater evapotranspiration
H	[L]	Optimal distance between root zone and water table. Phreatic zone irrigation should maintain this optimal distance.
H_z	[L]	Distance by which the water table is raised by phreatic irrigation
I	[L/T]	Average net infiltration rate in the month before the crop season
k_s	[L/T]	Saturated hydraulic conductivity
L	[−]	Eq. 32
M	[−]	Total crop solute uptake
n	[−]	Brooks and Corey soil hydraulic model parameter
N	[−]	Van Genuchten soil hydraulic model parameter
P	[L]	Crop water uptake originating from precipitation
q	[L/T]	Mean upwards flux into the root zone
s	[L]	Material coordinate (Eq. 15)
S	[L]	Crop water uptake originating from the recirculation of vadose zone stored moisture
t	[T]	Temporal coordinate
T	[T]	Duration of crop season
v	[L/T]	Flow velocity
V	[L]	$S - (ET_c - P)$
W	[L ²]	Width of dispersed zone of solute plume (Eq. 25)
z	[L]	Vertical spatial coordinate, $z = 0$ at water table
Z_g	[L]	Pre-irrigation groundwater depth
Z_r	[L]	Depth of the bottom of the root zone
α	[L]	Mechanical dispersivity
μ	[1/T]	Solute biodegradation rate
θ	[−]	Volumetric water content
θ_m	[−]	Mean volumetric water content in the vadose zone
θ_r	[−]	Residual volumetric water content
θ_s	[−]	Porosity
Θ	[−]	Relative saturation
Θ_m	[−]	Mean relative saturation in the vadose zone
ψ	[L]	Pressure head
ψ_a	[L]	Air entry pressure
ψ^*	[L]	Threshold pressure for maximum evapotranspiration

phreatic zone that occurs before the start of the crop season. The average vadose zone moisture content is approximately at equilibrium with the average infiltration rate (Salvucci and Entekhabi, 1994), such that $\frac{\partial S}{\partial t} = 0$ before the crop season starts, thus we obtain (Kim et al., 1996)

$$\frac{\partial S}{\partial t} = 0 = I - k_s \Theta_m^{3+\frac{2}{n}} \quad (3)$$

where I is the average net infiltration rate (precipitation minus runoff and evapotranspiration) in the month before the crop season, k_s is the saturated hydraulic conductivity, n is a parameter of the Brooks and Corey soil hydraulic model, and Θ_m is the mean degree of relative saturation in the vadose zone. Noting that the available vadose zone moisture content S can be expressed as

$$S = (\theta_s - \theta_r) Z_g \Theta_m \quad (4)$$

where θ_s is the porosity (or equivalently, the volumetric water content at saturation), Z_g is the pre-irrigation groundwater level, and θ_r is the residual saturation, we obtain

$$I - k_s \left(\frac{S}{(\theta_s - \theta_r)Z_g} \right)^{3+\frac{2}{n}} = 0 \# \quad (5)$$

which can be solved for S , yielding

$$S = (\theta_s - \theta_r)Z_g \left(\frac{I}{k_s} \right)^{\frac{n}{3n+2}} \# \quad (6)$$

If the natural groundwater level Z_g is too deep or if the soil is too coarse textured, some of the vadose zone moisture content cannot be delivered to the root zone by capillary rise. After phreatic irrigation begins, the water table is raised to a predefined target level. The moisture content associated with S is thereby pushed upwards to a level that is sufficiently shallow for the water to be delivered to the root zone through capillary rise. Therefore, regardless of how deep the natural groundwater level is, the entirety of the initial vadose zone moisture content becomes accessible to crops, under phreatic zone irrigation. For conditions representative of the Netherlands, vadose zone recirculation contributes about half towards compensating for a precipitation shortage (Kroes et al., 2018), even without interventions to raise the water table. Hence, under phreatic zone irrigation, recirculation of vadose zone moisture, which is not directly exposed to the irrigated contaminants, will contribute even more to crop water requirements.

2.3. Raising the water table

In unsaturated soils, the volumetric water content θ can be expressed in terms of the relative saturation Θ

$$\theta = \theta_r + (\theta_s - \theta_r)\Theta \# \quad (7)$$

For the Brooks and Corey water retention model, Θ is a function of the pressure head ψ : $\Theta(\psi) = 1(\psi \leq \psi_a)$

$$\Theta(\psi) = \left(\frac{\psi}{\psi_a} \right)^{-n} \#(\psi > \psi_a) \quad (8)$$

where ψ_a is the air entry pressure.

The rooting depth (depth of the bottom of the root zone) is a representative depth for calculating root-groundwater interactions (e.g. Perri et al., 2022; Kramer and Mau, 2020; Vervoort and van der Zee, 2008). For the actual evapotranspiration rate to be equal to the potential evapotranspiration rate e at all times even under stochastically and temporally fluctuating precipitation rates, the water table must be sufficiently close to the rooting depth to ensure that capillary fluxes are always sufficient to compensate for soil moisture shortfalls. This occurs when the distance H between the rooting depth and water table satisfies (Vervoort and van der Zee, 2008)

$$H \left[1 - \exp \left(-\beta \left[\left(\frac{H}{\psi_a} \right)^{-n} - \Theta^* \right] \right) \right]^{-\frac{1}{3n+2}} = \psi_a \left(\frac{e}{Ak_s} \right)^{-\frac{1}{3n+2}} \# \quad (9)$$

where Θ^* is the threshold root zone saturation beneath which the actual evapotranspiration rate is lower than the potential evapotranspiration rate. An analytical expression of the coefficient A based on soil properties is given by (Salvucci, 1993)

$$A = \left(1 + \frac{e}{k_s} \right)^{1-(3n+2)} \left[\frac{\pi}{(3n+2)\sin\left(\frac{\pi}{3n+2}\right)} \right]^{(3n+2)} \# \quad (10)$$

and for the coefficient β (Laio et al., 2001),

$$\beta = 4 + \frac{2}{n} \# \quad (11)$$

The saturation Θ^* corresponds to a suction pressure ψ^* that depends on the crop species. For example, $\psi^* = 3200\text{mm}$ for potatoes, sugar beets, and maize, $\psi^* = 2000\text{mm}$ for pasture, and $\psi^* = 5000\text{mm}$ for

wheat (Feddes and Raats, 2004). Putting these together yields

$$H \left[1 - \exp \left(- \left(4 + \frac{2}{n} \right) \left[\left(\frac{H}{\psi_a} \right)^{-n} - \left(\frac{\psi^*}{\psi_a} \right)^{-n} \right] \right) \right]^{-\frac{1}{3n+2}} = \left[k_s \left(1 + \frac{e}{k_s} \right)^{-3n-1} \right]^{\frac{1}{3n+2}} \left[\frac{\pi \psi_a}{(3n+2)\sin\left(\frac{\pi}{3n+2}\right)} \right] \# \quad (12)$$

which can be solved implicitly to obtain H .

2.4. Advective-dispersive tracer solute transport to the root zone

When the flux q is much smaller than the saturated hydraulic conductivity k_s of the soil $\frac{q}{k_s} \ll 1$, as is the case under capillary fluxes, the following small flux approximation applies for calculating the pressure head ψ at vertical position z at steady state (Parlange et al., 1990)

$$z \approx \int_0^\psi 1 \, d\psi' = \psi \# \quad (13)$$

where we use $z = 0$ at the water table. This results in a linear pressure profile with depth. Typical values of $\frac{q}{k_s}$ for capillary rise under evapotranspiration driven capillary rise are 0.0005 for sandy loam, 0.005 for silt loam, and 0.05 for clay (Salvucci, 1993). This means that the small flux approximation typically applies for most soil types during capillary rise, possibly with the exception of highly impermeable clays. Therefore, the relative saturation as a function of the vertical coordinate may be expressed as $\Theta(z) = 1(\psi \leq \psi_a)$

$$\Theta(z) = \left(\frac{z}{\psi_a} \right)^{-n} \#(\psi > \psi_a) \# \quad (14)$$

Under temporally fluctuating rainfall, although the soil moisture profile also experiences temporal fluctuations, the temporal mean soil moisture profile can be approximated with the steady-state soil moisture profile (Salvucci and Entekhabi, 1994). Furthermore, much of the soil moisture fluctuations occur near the topsoil, whereas the soil moisture content in below rooting depth fluctuates minimally (Salvucci and Entekhabi, 1994). Therefore, in the following calculations of solute transport from the water table to the root zone, we use this steady-state soil moisture profile to determine the average soil moisture content, which influences the extent of solute dispersion.

To describe solute transport in the vadose zone, it is necessary to first perform a change in coordinates. The following

$$s(z, t) = \int_0^z \theta dz - qt \# \quad (15)$$

transforms the spatiotemporal coordinate system (z, t) into a moving material coordinate system $s(z, t)$, where by definition the rising water front is located at $s(z, t) = 0$. The moving coordinate system is based on the principles of material coordinates, which assumes that all present soil moisture is perfectly displaced by a moving front, and which has been successfully validated against experimental evidence (Ellsworth and Jury, 1991; Bond and Phillips, 1990; Smiles, 2000).

The moving material coordinate system may be expressed as

$$s(z, t) = \left[\theta_r z + (\theta_s - \theta_r) \int_0^z \Theta(z) dz \right] - qt \# \quad (16)$$

where $\int_0^z \Theta(z) dz$ is the depth-averaged saturation of the soil already traversed by the rising solute front, given by

$$\int_0^z \Theta(z) dz = \psi_a \left(\frac{1}{1-n} \right) \left[\frac{z}{\psi_a} \left(\frac{z}{\psi_a} \right)^{-n} - n \right] \#(n \neq 1)$$

$$\int_0^z \Theta(z) dz = \psi_a \left[1 + \log \left(\frac{z}{\psi_a} \right) \right] \#(n = 1) \quad (17)$$

A special case of the analytical expression of $\int_0^z \Theta(z) dz$ is applicable when $n = 1$, because the general expression has an undefined result when $n = 1$.

The dispersion of solutes in the moving coordinate system is governed by the one-dimensional diffusion equation (Wilson and Gelhar, 1981)

$$\frac{\partial c}{\partial t} = (\theta^2 D)_{s=0} \frac{\partial^2 c}{\partial s^2} \# \tag{18}$$

where $c(z, t)$ is the concentration, and D is the hydrodynamic dispersion coefficient. Following recent research (Dou et al., 2022; Zhuang et al., 2021) which revealed that at low relative saturations $\Theta < 0.7$ (characteristic of capillary rise) the hydrodynamic dispersion coefficient D increases as the pore saturation increases, we define the hydrodynamic dispersion coefficient as

$$D = \theta \alpha v = \alpha q \# \tag{19}$$

where α is the mechanical dispersivity, v is the water flow velocity, and q is the volumetric flux. With this model, the hydrodynamic dispersion coefficient is independent of the flow velocity, in agreement with Vanderborght and Vereecken (2007), who found that D is independent of v in unsaturated soils except when flow is controlled by large inter-aggregate pores. As capillary rise mostly occurs within smaller intra-aggregate pores (Hird and Bolton, 2017; Gumbs and Warkentin, 1976), using $D = \theta \alpha v = \alpha q$ is justified. Hence, the mechanical dispersivity of our model has a different saturation dependence than the typical form used in popular numerical models such as HYDRUS and SWAP (Šimůnek et al., 2005; Kroes et al., 2017), which are general-use models for unsaturated zone flow and may be less accurate for specifically simulating solute transport through a limited subset of the available soil pore space (Mencaroni et al., 2021). Given that capillary fluxes should fully compensate for precipitation shortages under phreatic irrigation, the average upwards flux from the vadose zone to the root zone during the crop season is given by

$$q = \frac{ET_c - P}{T} \# \tag{20}$$

where T is the crop season duration.

The background concentration of solutes in the vadose zone moisture is initially c_b . For the CECs in (treated) wastewater, we assume $c_b = 0$. At the moment phreatic irrigation begins, we assume that the groundwater contains a uniform concentration c_e of contaminants that originate from the irrigated effluent. Thus, the interface between vadose zone moisture and groundwater initially constitutes a sharp solute front. As evapotranspiration consumes soil moisture, the solute front moves upwards with capillary fluxes, which causes contaminants to disperse around the solute front.

For a step change in concentration from 0 to c_e at the vadose zone and groundwater interface at $t = 0$, the initial and boundary conditions are

$$\begin{aligned} c &= 0, \quad z > 0, \quad t = 0 \\ c &= 1, \quad z < 0, \quad t = 0 \# \end{aligned} \tag{21}$$

where c is the dimensionless concentration normalized between c_b and c_e . The solute concentration is given by the solution

$$c(s, t) = \frac{1}{2} \operatorname{erfc} \left[\frac{s}{\sqrt{4W}} \right] \# \tag{22}$$

Here, W is the width of the dispersed zone of the solute plume and is a function of the dispersion parameters (Wilson and Gelhar, 1981).

$$W = \int_0^T (\theta^2 D)_{s=0} dt \# \tag{23}$$

where the upper domain of integration T applied here signifies that the solution applies to the situation at the end of the crop season. Note that at this stage, crop solute uptake has not yet been accounted for. The term $(\theta^2 D)_{s=0} = \theta^2 \alpha q$ is time-dependent as the solute front moves upwards over time, and this formulation of W cannot be analytically solved to yield closed-form solutions. However, Elrick et al. (1994) showed that under steady-state capillary rise, an accurate approximation can be achieved by substituting the spatially-averaged volumetric moisture content θ_m for θ in $(\theta^2 D)_{s=0}$. This yields

$$W = \int_0^T \theta_m^2 \alpha q dt = \theta_m^2 \alpha q T \# \tag{24}$$

Using Eq. 20 for q yields

$$W = \theta_m^2 \alpha (ET_c - P) \# \tag{25}$$

To calculate W , we need to first obtain the average volumetric moisture content θ_m , given by

$$\theta_m = [\theta_s - \theta_r] \Theta_m + \theta_r \# \tag{26}$$

where Θ_m is the spatially-averaged relative moisture content in the vadose zone, itself given by

$$\Theta_m = \frac{1}{H} \int_0^H \Theta(z) dz \# \tag{27}$$

where H is the distance between the root zone and the raised water table during phreatic zone irrigation, and $\int_0^H \Theta(z) dz$ is given by Eq. 17.

The total mass of solutes M that reaches the rooting depth is given by

$$M = \int_v^\infty c(s, t) ds = \left[\sqrt{\frac{W}{\pi}} \exp\left(-\frac{V^2}{4W}\right) - \frac{1}{2} \operatorname{Verfc} \left[\frac{V}{\sqrt{4W}} \right] \right] \# \tag{28}$$

$$V = S - (ET_c - P) = (\theta_s - \theta_r) Z_g \left(\frac{I}{k_s} \right)^{\left(\frac{n}{3n+2} \right)} - (ET_c - P) \# \tag{29}$$

Following the literature (e.g. Cornelissen et al., 2021; Calderón-Preciado et al., 2011; Šimůnek and Hopmans, 2009), we assume that crop solute uptake is passive as contaminants are present in trace concentrations, implying that crop solute uptake is equal to M . This is a reasonable assumption because CECs in the wastewater should not exceed trace concentrations to be assessed as safe for irrigation. If some CEC species are physiologically (partially) excluded from crop root uptake (Miller et al., 2016), this model would yield a conservative high-estimate of crop contamination, in line with our intentions to err on the side of safety.

2.5. Solute adsorption and biodegradation

An essential benefit of phreatic zone wastewater irrigation, as opposed to surface irrigation, is that CECs are subject to biogeochemical reactions on their way up towards the root zone. These biogeochemical reactions are adsorption, which slow the movement of solutes relative to the rising front of water, and biodegradation, in which soil microbes transform the CECs into possibly less ecotoxic product compounds, leading to lower overall crop solute exposure and uptake.

Commonly used models for the biogeochemical behavior of soil contaminants are linear equilibrium adsorption and first-order biodegradation. The linear adsorption model describes the adsorption behavior of CECs in soils well, particularly due to their presence in the soil in trace concentrations (Kodešová et al., 2015). Following Chrysikopoulos et al. (1990),

$$s_R(z, t) = R \left[\theta_r z + (\theta_s - \theta_r) \int_0^z \Theta(z) dz \right] - qt \# \tag{30}$$

$$W_R = \int_0^T R\theta_m^2 \alpha q dt = RW \# \quad (31)$$

where the subscript R implies that the solutions of s , W are relevant to biogeochemically reactive solutes.

We assume first-order biodegradation with a biodegradation rate of μ , which describes well the biodegradation of low concentrations of organic contaminants (Alexander, 1985; Birch et al., 2018). We also assume that biodegradation occurs only in the vadose zone between the water table and root zone, which is reasonable as biodegradation rates are typically much higher in the vadose zone than in the saturated zone due to higher microbe populations and oxygen concentrations for aerobic biodegradation (Borden and Bedient, 1986). This assumption is also conservative with respect to crop contamination risk, due to the implication that biodegradation of solutes stops once the solute reaches the root zone. Furthermore, we also assume that biodegradation occurs only for solutes in the aqueous phase, and not in the adsorbed phase, following experimental evidence (Beltman et al., 2008). For this model of biodegradation, the fraction of solute mass that reaches the root zone without being attenuated along the way is (Beltman et al., 2008; Jury and Gruber, 1989)

$$L = \exp \left(-S \frac{\sqrt{1 + \frac{4\mu\alpha\theta_m^2 T}{ET_c - P}} - 1}{2\alpha\theta_m^2} \right) \# \quad (32)$$

If there is reason to model biodegradation occurring in both the aqueous and adsorbed phase, the analytical solution can be found by multiplying the aqueous phase biodegradation rate μ by the retardation factor R (Jury and Gruber, 1989; Yang et al., 2016).

Hence, the dimensionless total crop solute uptake of biogeochemically reactive solutes M_R is

$$M_R = \left[\sqrt{\frac{RW}{\pi}} \exp \left(-\frac{V_R^2}{4RW} \right) - \frac{1}{2} V_R \operatorname{erfc} \left[\frac{V_R}{\sqrt{4RW}} \right] \right] \bullet \exp \left(-S \frac{\sqrt{1 + \frac{4\mu\alpha\theta_m^2 T}{ET_c - P}} - 1}{2\alpha\theta_m^2} \right) \# \quad (33)$$

$$V_R = RS - (ET_c - P) \# \quad (34)$$

If μ and R of a contaminant of interest are known, and acceptable levels of crop uptake are defined, then one could define permissible levels of contaminant concentrations c_e in the irrigation water. This allows for the design and management of phreatic zone irrigation systems, the evaluation of its feasibility, and regulation by authorities to be performed in a fast and general manner.

3. Sensitivity analysis

We characterize the sensitivity of crop uptake of tracer solutes M to changes in parameters $\{p_1, p_2, \dots, p_i\}$ through a local sensitivity analysis using the relative sensitivity function $U(p)$ (Boekhold, Van der Zee, 1991, Yang et al., 2016)

$$U(p) = \frac{\partial \log_{10} M_R}{\partial \log_{10} p} \# \quad (35)$$

where p is the parameter whose model sensitivity is being investigated. The relative sensitivity $U(p)$ may take on a range of $-\infty < U(p) < \infty$. If $U(p) < 0$, then an increase in p leads to a decrease in M , and if $U(p) > 0$,

then an increase in p leads to an increase in M .

For soils with textures ranging from sand to clay, the applicable range of parameters is approximately $0.5 < n < 3$, $0.2 < \theta_s < 0.5$, and $100\text{mm/day} < k_s < 1000\text{mm/day}$ (Kim et al., 1996). In the Netherlands, natural groundwater levels at sites with pipe drainage are typically at depths of $500\text{mm} < Z_g < 2000\text{mm}$. The value of α lies between 10 mm to 1000 mm, and would typically be around 50 mm – 100 mm for problems with a transport scale of around 1 m (Vanderborght and Vereecken, 2007). In the Netherlands, the nationally-averaged reference precipitation shortage during the crop season has an across-year median of 100 mm, and a two standard deviation high around 300 mm, according to the Dutch meteorological institute (KNMI). Parameter values used in the base case of the sensitivity analysis are chosen to be typical for an agricultural field requiring irrigation in the Netherlands, for an intermediate (loamy) soil type. Therefore, we use $\theta_s = 0.35$, $\theta_r = 0.01$, $T = 150\text{days}$, $I = 1.5\text{mm/day}$, $ET_c - P = 200\text{mm}$, $k_s = 100\text{mm/day}$, $n = 1$, $\psi_a = 300\text{mm}$, $\psi^* = 3000\text{mm}$, $e = 2\text{mm/day}$, $Z_g = 1500\text{mm}$, $\alpha = 100\text{mm}$, $R = 1$, $\mu = 0.01/\text{day}$ for the base case.

From Eq. 6, the vadose zone stored moisture content S can be fully characterized by plotting S/Z_g as a function of I/k_s and n on a two-dimensional graph. As shown in Fig. 2a, S/Z_g increases as I increases, as k_s decreases, as n decreases, and as Z_g increases. Since the domain of parameter values used in the contour plots is similar to the domain of parameter values in the Netherlands, the results in Fig. 2a allow us to generalize that $0.1Z_g < S < 0.2Z_g$ in general in such a climatic zone. Numerical simulations reveal that for a fixed crop type (i.e. ψ^* fixed), H/ψ_a can essentially be characterized as a function of k_s/e and n (Fig. 2b), with only negligible variations in H/ψ_a as ψ_a is varied. Fig. 2b shows that H increases as n decreases, as k_s increases, as e decreases, and as ψ_a increases. Hence, we have performed a dimensionless analysis of S and H . The optimal distance between the root zone and water table, H , is therefore between 1.5 and 4 times of ψ_a for most combinations of k_s/e and n (Fig. 2b).

For an unreactive tracer in the base case $M = 30$ instead of zero, even though the vadose zone stored moisture was enough to compensate for the entire precipitation shortage ($S \approx ET_c - P \approx 200\text{mm}$), because of contaminant dispersion in the vadose zone. In contrast, if it was assumed that the entire precipitation shortage was fulfilled with undiluted wastewater at the original concentration c_e , the corresponding root solute uptake would be 200. Hence, accounting for the role of the water stored in the vadose zone in fulfilling crop evapotranspiration requirements, and the dispersion of contaminants within the vadose zone during capillary rise, is important for assessing the quality of the water taken up by the plants. For a reactive solute in the base case with $R = 2$ and $\mu = 0.01/\text{day}$, we find $M_R = 0.01$. These values of the biogeochemical reaction parameters are similar to those used in Tang et al. (2023) for carbamazepine, and crop uptake of carbamazepine was found to be negligible in that study, in agreement with this analysis. This shows that even a relatively small retardation factor and biodegradation rate can lead to large reductions in crop contamination risk, in agreement with Tang et al. (2023).

For a solute (adsorbing or non-adsorbing) that does not undergo biodegradation, $M_R(\mu = 0)$ can be fully described with two lumped parameters: V_R and RW . Fig. 3a shows that $M_R(\mu = 0)$ increases as V_R decreases, and as RW increases. At large V_R , changes to RW have much

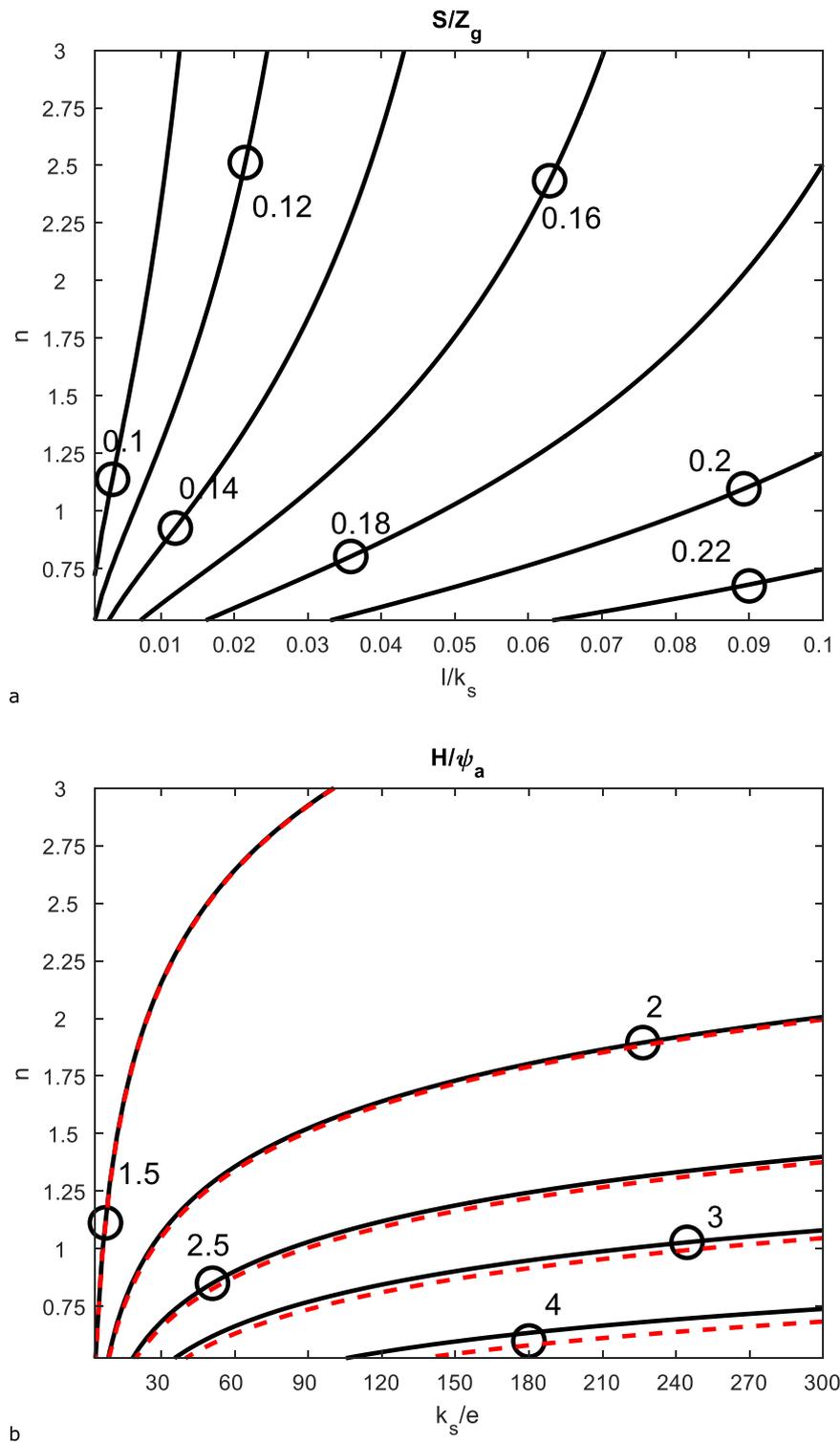


Fig. 2. Contour plots of (a) S/Z_g as a function of I/k_s and n . (b) H/ψ_a as a function of k_s and n . The black solid lines are for $\psi_a = 200\text{mm}$, and the red dashed lines are for $\psi_a = 400\text{mm}$.

larger relative effects on $M_R(\mu = 0)$ than at small V_R . In other words, changes to the width of the solute plume front RW have a larger effect on $M_R(\mu = 0)$ when the solute plume front is far from the root zone. At small RW , changes to V_R have a much larger relative effect on $M_R(\mu = 0)$ than at large RW . This is because the concentration gradient is larger when the width of the plume front is small.

It is also possible to characterize $M_R(\mu = 0)$ with the three lumped parameters RS , $ET_c - P$, and $\theta_m^2\alpha$. Fig. 3b shows that depending on the

values of RS and $ET_c - P$, $M_R(\mu = 0)$ may increase by multiple orders of magnitude, or hardly increase, when $\theta_m^2\alpha$ is increased by an order of magnitude. If $ET_c - P$ is smaller than RS , then $M_R(\mu = 0)$ is very sensitive to $\theta_m^2\alpha$, as solute concentrations in the vadose zone moisture taken up by crops depend heavily on the effective solute dispersivity $\theta_m^2\alpha$. However, if $ET_c - P$ is much larger than RS , then $M_R(\mu = 0)$ is not sensitive to $\theta_m^2\alpha$, as crops are taking up solutes from the groundwater at nearly wastewater concentrations, which means that solute dispersion becomes

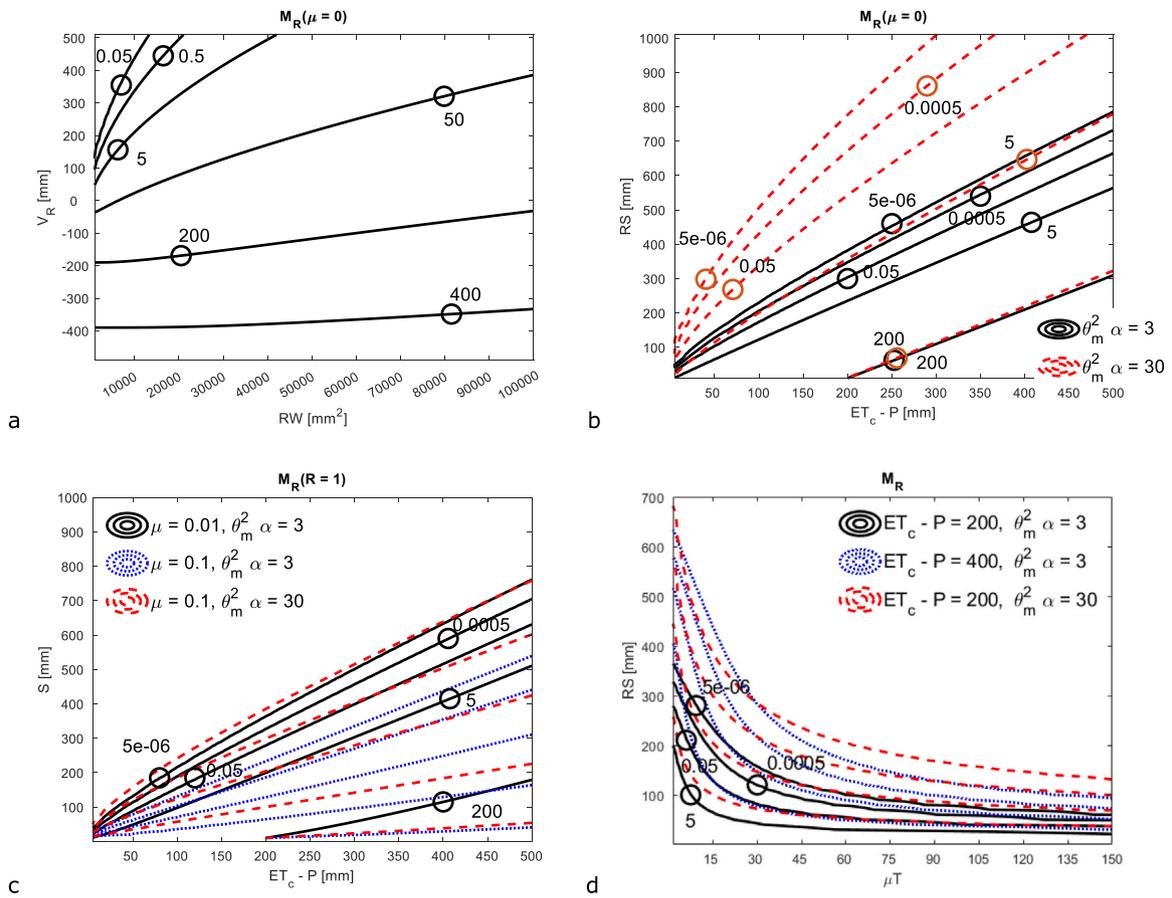


Fig. 3. Contour plots of M_R as a function of (a) V_R and RW when $\mu = 0$, (b) RS and $ET_c - P$ when $\mu = 0$, (c) S and $ET_c - P$ when $R = 1$, and (d) RS and μT , and how they change when the other remaining parameters of the model are varied (see legend).

mostly irrelevant.

Fig. 3c shows $M_R(R = 1)$, the root solute uptake for a biodegrading but non-retarding solute. This case is fully characterized by four (lumped parameters) S , $ET_c - P$, μ , and $\theta_m^2 \alpha$. As the biodegradation rate μ increases, $M_R(R = 1)$ decreases as expected. As the effective solute dispersivity $\theta_m^2 \alpha$ increases, $M_R(R = 1)$ increases. Similarly to the case of $M_R(\mu = 0)$, the sensitivity of $M_R(R = 1)$ to $\theta_m^2 \alpha$ is larger when S is larger and $ET_c - P$ is smaller. Fig. 3c also shows that when $M_R(R = 1)$ is small, increasing both the biodegradation rate μ and the effective solute dispersivity $\theta_m^2 \alpha$ by 10 times may actually lead to increased $M_R(R = 1)$, which means that even though M_R may be more sensitive to $\theta_m^2 \alpha$ at small M_R and more sensitive to μ at large M_R .

The effects of the crop season duration T and the biodegradation rate μ are entirely contained within the biodegradation term L . As evident in Eq. 32, any combination of μ and T that yields the same value of the product μT leads to identical values of L . We calculated M_R with varying values of RS , μT , $ET_c - P$, and $\theta_m^2 \alpha$, and plotted the outcomes in Fig. 3d. Here, it is shown that the sensitivity of M_R to the various parameters is small when μ is small, but the sensitivity is large when μ is large (i.e. the distance between contour lines is smaller). Therefore, when μ is large, uncertainties in other model parameters have larger adverse effects on accurately predicting M_R . Uncertainty in μ itself is large in field conditions, as it may vary across multiple orders of magnitude due to differences in soil biogeochemical conditions (Nham et al., 2015). Hence, estimates of the crop contamination risk by highly biodegradable solutes with large μ is unlikely to be accurate nor meaningful for field situations. Therefore, crop contamination risk under phreatic zone wastewater irrigation should in general be quantified based on crop exposure risks to non-biogeochimically reactive tracers contained within the effluent.

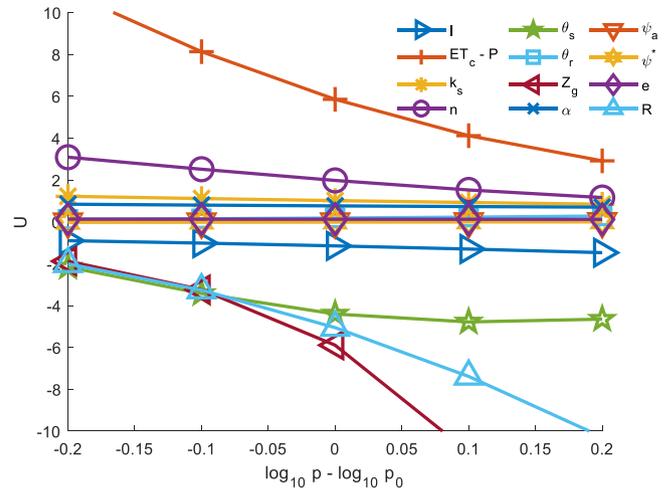


Fig. 4. The relative sensitivity of M to changes in parameters, as a function of $\log_{10} p / p_0$, where p_0 refers to the reference (base case) value of the parameter.

We performed a local sensitivity analysis using the relative sensitivity function $U(p)$, for a non-biodegrading solute ($\mu = 0$), with other parameters at base case values (Fig. 4). Evidently, the parameters that most affect M_R are I , θ_s , n , Z_g , $ET_c - P$, R ; these are the parameters that comprise the lumped parameter V_R (Eq. 34). Thus, aside from the retardation factor, the parameters that affect M most are the parameters that affect how much of the crop water uptake originates from the vadose zone stored moisture S and groundwater evapotranspiration G .

Fig. 4 also reveals that an increase in the parameters θ_r , $ET_c - P$, n , k_s , α , e causes M_R to increase, whereas an increase in θ_s , Z_g , I , R causes M to decrease. The parameter ψ_a has negligible effect on M_R . This is because ψ_a only affects M_R through θ_m . However, θ_m is fully determined by the ratio H/ψ_a , which is essentially independent of ψ_a (Fig. 2b). Therefore, changes in ψ_a do not affect θ_m and thus also do not affect M_R . The crop-related hydrological parameters ψ^* and e also have minimal impact on M_R , meaning that crop species only affect the crop contamination risk by tracers through the precipitation shortage $ET_c - P$. Any additional effects of biodegradation (and the parameters μ and T) are evident in the biodegradation term L (Eq. 32), and also in the analysis of Fig. 3d in the previous paragraph.

4. Analytical model concept validation

As explained throughout the derivation of our analytical model, all of the physical concepts and mathematical equations we applied have been sourced from the literature, where they have been shown to be validated against experimental and observational data. The main novelty of the model we introduce, and a key reason for its simplicity, is our concept of estimating crop contamination based on the overall crop season water balance of the root zone. The concept in other words is: since the location of contaminant plume's front is determined by the crop season water balance of the root zone, detailed time series of daily or hourly precipitation and evapotranspiration data is unnecessary for estimating contaminant fate. This would be particularly useful for coarse assessments of crop contamination risks across a large spatial scale, with low data and computational requirements.

To validate our concept of using the root zone water balance to estimate crop contamination, we first compared sampled root zone tracer concentrations against the cumulative crop season precipitation shortage over four years (four crop seasons) of a field experiment. Briefly described, we conducted a field experiment of the subsurface irrigation and drainage system on a 58500 m² field in Haaksbergen, the Netherlands, where maize crops for livestock fodder was grown (see Fig. 1a for a conceptual illustration of the field experiment, and see Tang et al. 2023 and Narain-Ford et al. 2022 for further details of the experimental setup). Treated wastewater from a domestic wastewater treatment plant was fed into the subsurface irrigation and drainage system, which operated during the crop season (May to September) as an irrigation and drainage system, and the rest of the year as a drainage system. The chloride to bromide ratio (Cl:Br) was used as a tracer, and

soil water samples from 0.6 m depth directly above an irrigation pipe were analyzed for root zone tracer concentrations. At the study site, Cl:Br of environmental freshwater is typically between 100 – 300, and Cl:Br of the treated wastewater is typically between 800 – 1200. As no irrigation occurs outside of the crop season, and as precipitation excesses of around 300 mm to 400 mm occur during the non-crop seasons, the root zone solute concentrations are reset to background environmental concentrations by the start of every new crop season except if a severe drought occurs during the preceding year (Tang et al., 2023; Narain-Ford et al., 2022). A severe drought occurred at the study site in 2018, explaining the relatively elevated Cl:Br in 2018 and the subsequent year 2019 (Fig. 5).

As Fig. 5 shows, there appears to be a substantial correlation between the within-crop-season cumulative precipitation shortage and the root zone tracer concentration. This observed correlation reinforces the validity of this present study, where we use the within-crop-season root zone water balance (parameters $ET_c - P$ and S) as substitutes for detailed weather time series in estimating crop contamination risks. This correlation is nonlinear due to the nonlinear solute concentration profiles generated under hydrodynamic dispersion (Tang and Rijnaarts, 2023), which suggests that the analytical solute uptake model we introduce would better describe root solute uptake risks than a simple linear correlation analysis between ET-P and root solute uptake. Although the field measured concentrations in 2017 appear relatively more detached from the ET-P curve than in other years, this could be explained by the large extent of plume front oscillations resulting from highly variable weather (discussed in more detail below), as can be seen from the fluctuating (saw-toothed-shaped curve in 2017 of the) cumulative precipitation shortage (Fig. 5).

Additionally, in Tang et al. (2023) we created a numerical model of contaminant fate (validated against the field experiment) that uses detailed daily weather data (precipitation and actual evapotranspiration; see Tang et al., 2023 for more details), as opposed to the cumulative precipitation shortage used in the present study's analytical model. To further validate the analytical model introduced in the present study, we ran that numerical model with 30 years of historical daily weather data (Twenthe weather station, the Netherlands) from the experimental site. Aside from the newly introduced 30-year weather data, the construction and parameters of the numerical model are identical to the 'base case' scenario of Tang et al. (2023), with the irrigated wastewater containing a generic tracer at a constant dimensionless concentration of 1. We then applied the analytical model of this study to estimate root solute uptake of an unreactive tracer (Eq. 28), and compared its output with the 30 year numerical simulations. The parameter values for the analytical model were chosen to approximate the circumstances of the numerical simulations, and are $\theta_s = 0.42$, $\theta_r = 0.01$, $T = 150$ days, $I = 1.5$ mm/day, $k_s = 500$ mm/day, $n = 2$, $\psi_a = 80$ mm, $\psi^* = 3000$ mm, $e = 2$ mm/day, $\alpha = 200$ mm/ θ_m , $c_e = 0.19$, $Z_g = 1000$ mm. The value of $c_e = 0.19$ for the one-dimensional analytical originates from the observation that a dimensionless tracer input concentration of 1 from the irrigation pipes leads to an average dimensionless tracer concentration of 0.19 in the saturated zone, under the assumptions and parameters of the two-dimensional numerical model. Soil hydraulic parameter conversions between the numerical model (Van Genuchten parameters) and analytical model (Brooks and Corey parameters) were performed using the equivalence relationships of Morel-Seytoux et al., (1996).

An oscillating solute plume front due to fluctuating weather causes the effective plume dispersal width W (Eq. 25) to be larger than the value applicable to steady-transport (Cirkel et al., 2015), without significant effects on the mean solute transport and breakthrough behavior (Elhanati et al., 2023). This issue can be approached by using a larger value of W , ignoring erratic weather, and considering only average fluxes, which has shown to be sufficient in producing accurate estimates of solute transport to the root zone from underlying groundwater (Stofberg et al., 2017). In the numerically simulated scenarios discussed above, we find that the contaminant plume front tends to oscillate

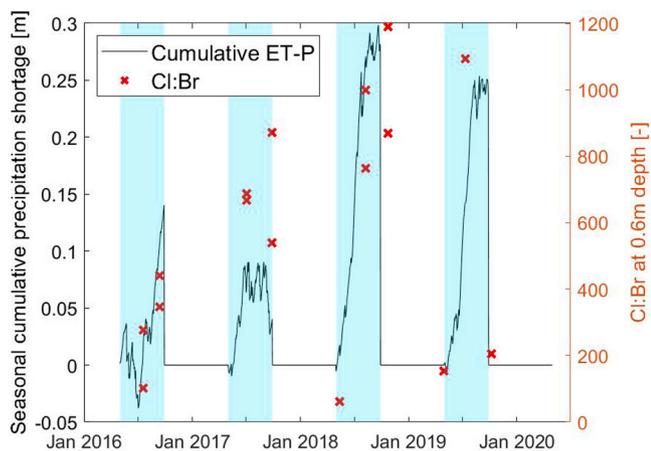


Fig. 5. Timeseries of the seasonal cumulative precipitation shortage against the Cl:Br of soil water samples. The time periods highlighted in cyan refer to the crop season. The calculated cumulative precipitation shortage ET-P are reset to zero before each new crop season, because the root zone is essentially contaminant-free at the start of every crop season except if a severe drought occurs during the non-crop season (Tang et al., 2023; Narain-Ford et al., 2022).

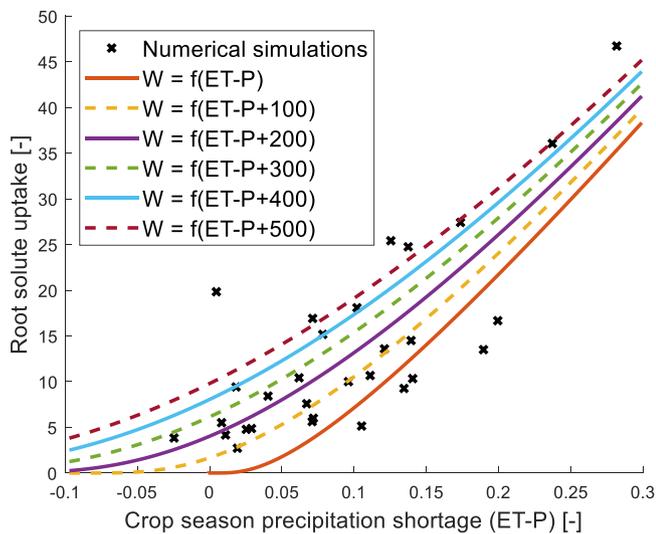


Fig. 6. The root solute uptake as calculated using the numerical model of Tang et al. (2023) with 30 years of daily weather data, as compared to the analytical model of this study (Eq. 28). The $f()$ symbols in the legend stand for ‘function of’. Each point in the scatter plot refers to one of the 30 years simulated. The plotted lines show the analytically predicted root solute uptake under a range of contaminant plume front oscillations between 0 mm to 500 mm.

around 200 mm in absolute displacement, in response to fluctuating weather. Therefore, we calculated the analytical model, but substituted $(ET - P + \text{oscillation displacement})$ in place of $(ET - P)$ when calculating W (Eq. 25). In Fig. 6, we show the analytical model outcomes when plume front oscillations ranging between 0 mm to 500 mm in absolute displacement are taken into account. The cloud of numerical simulation data is well captured around this range of analytical model outcomes. Therefore, the extent of oscillatory movement may be treated as a stochastic parameter in order to obtain a range of possible outcomes with the analytical model.

In summary, even though the analytical model 1) requires only the cumulative precipitation shortage of the crop season but not daily weather data timeseries, and 2) is a one-dimensional model that ignores the non-vertical spatial aspects of solute transport, it can well explain the results of the much more complex numerical model of Tang et al. (2023). Therefore, the analytical model introduced in this study agrees with the field-calibrated numerical model of Tang et al. (2023), in their descriptions of how root solute uptake is affected by the root zone water balance. The analytical model thus successfully captures the main processes that determine crop contamination risks, under the assumptions of the present study. Consequently, the analytical model is valid for coarsely estimating crop contamination risks, and for performing sensitivity analyses of crop contamination risks in response to parameter variations. Given the novelty of intentionally irrigating wastewater beneath the water table to utilize the vadose zone as a contaminant buffer, this is the first study to analytically model this irrigation method.

5. Discussion

5.1. Analytical model limitations

A key simplifying assumption of this study is that crop water requirements are satisfied from the three compartments in the order of preference $P \rightarrow S \rightarrow G$. Therefore, in the model, the vadose zone soil moisture and groundwater do not contribute to evapotranspiration during the crop season, unless $ET_c > P$. In terms of crop solute uptake, the effects of this simplification are that the duration that solutes experience biodegradation may be slightly overestimated. This is a minor source of error, considering that the biodegradation rate of any

particular reactive solute may vary across five orders of magnitude under various field conditions (Nham et al., 2015). This implies that the biodegradation rate, even if quantified in a laboratory setting, contributes more uncertainty to solute fate than the time available for biodegradation. Furthermore, this does not affect the root solute uptake estimates for the most hazardous solute types: persistent solutes that do not biodegrade in the soil (Narain-Ford et al., 2022).

Another simplification of the model is that it assumes a soil with uniform hydraulic properties throughout its depth, which may not be realistic. Nevertheless, soil hydraulic properties corresponding to the average soil texture can be used in our homogeneous analytical model to accurately estimate the field-scale mean crop solute uptake in heterogeneous soils. This is because even strong soil heterogeneity (including heterogeneity in both the horizontal and vertical directions) does not change the mean crop solute uptake, although it significantly affects solute concentrations and masses in the saturated zone (Tang et al., 2023). Furthermore, in heterogeneous soils, amongst all the various compartments of solute fate (e.g. crop uptake, leaching to confined aquifer, discharge within phreatic aquifer), crop solute uptake outcomes have by far the smallest relative variation under soil heterogeneity (Tang et al., 2023). This is because crop solute uptake is controlled to a large extent by the root zone water balance, and this property is fundamental the analytical model created in this study.

5.2. The role of irrigation method in crop contamination risks

The risks of crop contamination under wastewater irrigation also depend on the irrigation method. If wastewater is applied with traditional (near)surface irrigation techniques, such as surface irrigation or root zone drip irrigation, the root zone will be directly exposed to contaminants as there is no vadose zone buffer separating the wastewater from the root zone. The CECs in the wastewater will therefore not be attenuated through retardation and biodegradation in the vadose zone before arriving in the root zone. In contrast, phreatic zone wastewater irrigation retains rainwater in the root zone, keeping it relatively contaminant-free, but introduces CECs directly into the groundwater. Therefore, from the perspective of crop contamination risk under wastewater irrigation, phreatic zone irrigation leads to significantly lower risks of crop contamination than (near)surface irrigation methods. Furthermore, during phreatic zone wastewater irrigation, crops situated midway between the irrigation pipes are exposed to significantly less CECs than crops situated directly above irrigation pipes, because the irrigated solutes spread slowly to the groundwater and soil midway between pipes (Tang et al., 2023; Narain-Ford et al., 2022). This means that the contamination of crops not located directly above drains, and also the mean or total field-scale crop contamination risk, will be overestimated by our model. Hence, our model conservatively underestimates the reduction in crop contamination risk achieved by irrigating wastewater into the phreatic zone compared to conventional irrigation techniques, and can serve as a baseline for assessing the potential risk reduction achievable by adopting phreatic zone irrigation.

A similar irrigation system that has been previously studied is the “deep subsurface drip irrigation” technique of Bern et al. (2013). In their experiment, Bern et al. (2013) used sodic wastewater to irrigate crops through pipes buried 92 cm beneath the soil surface, below the root zone and above the water table. The contaminant of interest in their study is Na^+ ions. If the pipes are situated between the water table and root zone, then there is less contaminant-free water available for plant uptake stored in the vadose zone between the pipes and the root zone, than if the pipes were situated beneath the water table. The moisture stored in the vadose zone is substantial (Kroes et al., 2018), and would make a significant contribution in limiting crop contamination risks. In the context of our study, this means that S is smaller when the pipes are located above the water table. Nevertheless, the experimental findings of Bern et al. (2013) reveal that after six years, hardly any Na^+ reaches the root zone despite the small S , because the Na^+ ions experienced

significant retardation (Bern et al., 2013). Indeed, Na^+ ions exhibit especially large adsorption and retardation due to cation exchange when advection velocities are small (Zhen et al., 2016), such as during capillary driven flow. The experimental results of Bern et al. (2013) therefore agree with our study, where we show that solute retardation is instrumental in reducing crop contamination risks.

Another difference between phreatic zone irrigation and other subsurface irrigation methods, is that if subsurface irrigation occurs in the vadose zone (where fluxes are primarily vertical) instead of in the phreatic zone (where lateral fluxes from regional groundwater flow are significant), the contaminants are more likely to be contained within the irrigated field's subsurface, and may accumulate in the long term. This adverse effect is most likely to occur with contaminants that exhibit high retardation, which may be slow to leach towards the phreatic zone. However, as discussed in the previous paragraph, wastewater containing strongly retarding contaminants is also the type of wastewater most suitable for vadose zone drip irrigation with respect to crop contamination risks, because they are least likely to rise to the root zone. Therefore, wastewater with mobile and immobile contaminants are both not ideal for irrigation into the vadose zone. These issues are mitigated by irrigating into the phreatic zone: mobile contaminants are less likely to reach the root zone due to the larger separation distance and dilution by groundwater, whereas immobile contaminants are less likely to accumulate beneath the irrigated field because of advection by regional groundwater fluxes. It has already been shown, using numerical simulations (Tang et al., 2023) and field experimental evidence (Narain-Ford et al., 2022), that when irrigating wastewater through the phreatic zone, contaminants do not accumulate in the root zone or crops, as contaminant levels there reset to near background levels before the onset of every subsequent crop season, except in exceptional years with extremely large precipitation shortages. Furthermore, when irrigating through the phreatic zone, contaminant dilution by groundwater mitigates possible adverse effects of wastewater irrigation on soil hydraulic properties (Assouline et al., 2020). Hence, as contaminants are less likely to remain in the subsurface of the irrigated plot when wastewater is irrigated beneath the water table, uncertainties in the (long-term) biogeochemical behavior of various contaminant species in the root zone soil (e.g. Singh, 2021; Ruan et al., 2023; Lyu et al., 2022) will become less of an obstacle to the wider and more intensive adoption of wastewater irrigation.

Although wastewater irrigation has a long history (Zhang and Shen, 2019), and a large body of scientific literature (Hashem and Qi, 2021), a knowledge gap remains regarding the effects of long-term wastewater irrigation and the long-term effects of wastewater irrigation (Singh, 2021). These uncertainties are largely due to the differing biogeochemical and accumulating behavior in the crop-soil system and regional groundwater, of the various contaminants that may be present in the wastewater used for irrigation (e.g. Simhayov et al., 2023; Lyu et al., 2022; Ruan et al., 2023; Natasha et al., 2021; Liu et al., 2020; Sunyer-Caldú et al., 2023; Dang et al., 2019). Possible solutions to reduce long-term risks and uncertainties include stricter thresholds on the contaminant content of the irrigated wastewater or the use of hyper-accumulators to remediate polluted soils (Minhas et al., 2022). However, these options, respectively, limit the quantity of wastewater that can be reused, and may necessitate the farming of economically unproductive crops. We show that wastewater irrigation through the phreatic zone is associated with a smaller risk and uncertainty regarding long-term crop and root zone soil pollution, compared to other forms of surface or subsurface wastewater irrigation where water is applied above the water table. This is because the perpetual mobility of the groundwater reduces the risk that contaminants migrate to and accumulate in the root zone and vadose zone of an intensively cultivated soil. This in turn implies that the biogeochemical behavior of contaminants in the root zone soil, which is highly uncertain and thus responsible for a large degree of the uncertainty concerning long-term soil contamination risks, becomes less of a determining factor of long-term root zone

pollution. This comes, however, at the cost of reduced water-use efficiency.

5.3. Water use efficiency and wider environmental implications of phreatic zone irrigation

During phreatic zone irrigation, the water table has to be raised by some distance H_z , to maintain a distance of H between the root zone and water table. The irrigation flux F necessary to maintain the water table at the target height is equal to the rate of total water losses from the phreatic zone to lower aquifers and to the adjacent hydrological catchments. Generally, F is given by a function of the form

$$F = \frac{H_z}{B} \quad (36)$$

where B is an empirical parameter that depends on the hydraulic resistance of the surrounding subsurface environment (Di Ciacca et al., 2019; De Lange, 1999). Therefore, as the natural groundwater table depth Z_g becomes larger, the water usage of the irrigation method can be expected to increase proportionally, and its water use efficiency will decline accordingly.

The large irrigation volumes required implies the injection of possibly large amounts of contaminants into the subsurface, which may result in groundwater contamination. These contaminants released into the groundwater may either biodegrade gradually, or remain mostly within the phreatic zone if not biodegraded, until they are transported and eventually discharged to surface water (Tang et al., 2023). In the case of our experimental site, only relatively small amounts of contaminants are expected to seep to underlying confined aquifers (Narain-Ford et al., 2022; Tang et al., 2023), where they may continue to be attenuated (Aronson and Howard, 1997; Scow and Hicks, 2005; He et al., 2016).

Despite the environmental contamination risks, phreatic zone wastewater irrigation may nevertheless provide a superior alternative to typical methods of wastewater disposal. If not reused for purposes such as irrigation, a large amount of (treated) wastewater (e.g. from domestic or industrial wastewater treatment plants) would be discharged to surface water (e.g., rivers) and wasted. During dry crop seasons, when agricultural water demand is large and surface water bodies are dry, treated wastewater discharged to surface channels may be collected by farmers for irrigation at undiluted contaminant concentrations (Beard et al., 2019). Under phreatic zone irrigation, the soil and phreatic zone acts as a bioreactor for an additional step in the wastewater treatment process, before the effluent ultimately reaches other compartments of the environment, such as surface water or deeper groundwater. Nevertheless, this knowledge should be used within a full environmental risk assessment, in which all possible contamination routes are being considered for the specific geohydrological and climatic conditions of a site.

5.4. Future development of phreatic zone irrigation with wastewater

In this study, we considered the abiotic factors that influence crop exposure and uptake of solutes contained within the irrigated effluent. Biotic factors such as crop physiology and the soil microbial community may also affect crop solute uptake (Christou et al., 2019). The biotic factors that affect crop contamination risk are expected to be different when irrigating wastewater into the soil surface, root zone, vadose zone, or phreatic zone. For example, this is because the microbial community that affect contaminant biodegradation differ with soil depth, and differ in rhizosphere versus non-rhizosphere soil (Shi et al., 2022; Miller et al., 2016). Indeed it has been shown that root exudates and rhizosphere microbial communities may enhance contaminant bioremediation, and this could be optimized through the choice of plant species (Pilon-Smits, 2005), including many common crop plants such as wheat, maize, and

flux (Anderson et al., 1993). Hence, the effects of biotic factors on crop solute uptake should be researched in more detail. Nevertheless, the biotic factors that determine crop root interactions with the irrigated contaminants can only come into play if and when the solutes actually reach the root zone. Therefore, the abiotic transport factors that govern the transport of contaminants to the root zone, which we have modelled in this study, are the primary determinants of crop contamination risks.

The long-term effects of wastewater irrigation on soil functions should also be studied, in order to evaluate the sustainability of the practice (Cornelissen et al., 2021). Irrigation with wastewater may gradually alter the physical and biogeochemical characteristics of the soil. Soil quality may decline in the long term under wastewater irrigation, though the severity and potential remediation and mitigation methods are still under debate (van de Craats et al., 2020; Ibrahim et al., 2022; Jahany and Rezapour, 2020; Chaganti et al., 2021; Avishai et al., 2017; Leuther et al., 2019), and may depend on soil type, microbial ecology, and the chemical composition of the wastewater. If the soil physical and biogeochemical properties change over time, then so will the risk of crop contamination under phreatic zone wastewater irrigation. Future research into long-term changes to soil properties due to wastewater irrigation may provide further insight into the appropriate soil types, wastewater quality, and irrigation methods for reusing wastewater in agriculture.

The potential benefits of irrigation with wastewater are not limited to freshwater conservation. Wastewater irrigation could stabilize soil biogeochemical cycles (Santos et al., 2023), and could increase soil fertility as wastewater may contain readily-bioavailable macronutrients, micronutrients, and organic carbon, which are essential for optimal crop growth (Ofori et al., 2021). Accordingly, wastewater irrigation has been reported to increase crop yields compared to freshwater irrigation (Hassanli et al., 2010). With subsurface irrigation and controlled drainage, nutrient losses due to percolation from the root zone could also be reduced (Bonaiti and Borin, 2010). Furthermore, just as CECs present within the wastewater are advected and dispersed towards the root zone, so will nutrients in the wastewater. Microbes in the soil may also facilitate the transformation of nutrients into more bioavailable forms (Melia et al., 2017). Hence, solute transport from the irrigated wastewater to the root zone may become desirable, if the nutrient content of the irrigated effluent outweighs the risks posed by its CEC content. However, recent research has suggested that the economic value of wastewater irrigation is only minorly affected by the value of the nutrients contained within (Mainardis et al., 2022). Furthermore, the nutrient quantities and ratios in wastewater likely differ from crop-specific nutrient requirements (Urbano et al., 2017), meaning that simultaneously applying other methods of fertilization may still be necessary. Therefore, using wastewater as fertilizer should not be the main aim of water reuse. Nevertheless, understanding the fertigrative potential of wastewater irrigation in relation to its crop and environmental pollution risks has become a subject of interest (Chojnacka et al., 2020). In this regard, the risks and potentials of phreatic zone wastewater irrigation will likely differ from those of other irrigation methods.

6. Conclusion

In this study, we have derived a simple analytical model to determine crop solute uptake under phreatic zone wastewater irrigation, a new irrigation technique. The parameters that contribute most towards the capacity for moisture stored in the vadose zone to fulfil the crop water requirements, such as the crop-season precipitation shortage, groundwater depth, and soil volumetric water content at saturation, have the greatest impacts on crop contamination risk, especially for biogeochemically unreactive solutes. This shows that phreatic zone irrigation, which aims to compensate for crop-season precipitation shortages with the (relatively contaminant-free) moisture stored in the vadose zone, works as intended in terms of both mechanisms and outcomes. For biogeochemically reactive solutes, the biogeochemical parameters

(retardation factor and biodegradation rate) also significantly affect crop contamination risks. Crop solute uptake is highly sensitive to the mechanical dispersivity when crop water uptake is primarily fulfilled by recirculated vadose zone moisture, but relatively insensitive when crop water uptake is primarily fulfilled by groundwater evapotranspiration. Crop contamination risks in the former case cannot be characterized by non-spatially-explicit models of crop and soil contamination (e.g. Cornelissen et al., 2021). The added value of using the model introduced in this study is thus especially significant when the volume of vadose zone stored moisture is substantial, which also is the ideal scenario for applying phreatic zone wastewater irrigation.

The primary advantage of phreatic zone irrigation, over other irrigation techniques, is that crops are less exposed to the contaminants present in the wastewater. Furthermore, the risks and associated uncertainties of long-term accumulation of contaminations in the root zone soil is decreased compared to other irrigation techniques, as irrigating beneath the water table allows for flowing groundwater to dilute the contaminants and transport them away from the irrigated plot. This implies that knowledge gaps and uncertainties in the biogeochemical and accumulation behavior of various contaminant species in root zone soil become less important in determining crop contamination risks. The feasibility of this irrigation system, from the perspective of crop pollution and irrigation water requirements, can be broadly assessed at a wide geographic scale using the simple analytical model derived in this study, and geospatial hydrogeological and climate data. This study may serve to further inform the recently begun efforts of the EU and other regional governments to stimulate and regulate wastewater reuse, particularly with respect to relatively new irrigation techniques such as phreatic zone wastewater irrigation. Hence, within the framework of the environmental risk assessments that accompany the planning of wastewater reuse in agriculture, the analytical model introduced here may serve as a tool for rapid and broad assessment of the suitability of phreatic zone wastewater irrigation in relation to crop contamination risks, that does not require computationally-intensive numerical simulations.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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